Supporting Information

Discordant Nature of Cd in GeTe Enhances Phonon Scattering and Improves Band Convergence for High Thermoelectric Performance

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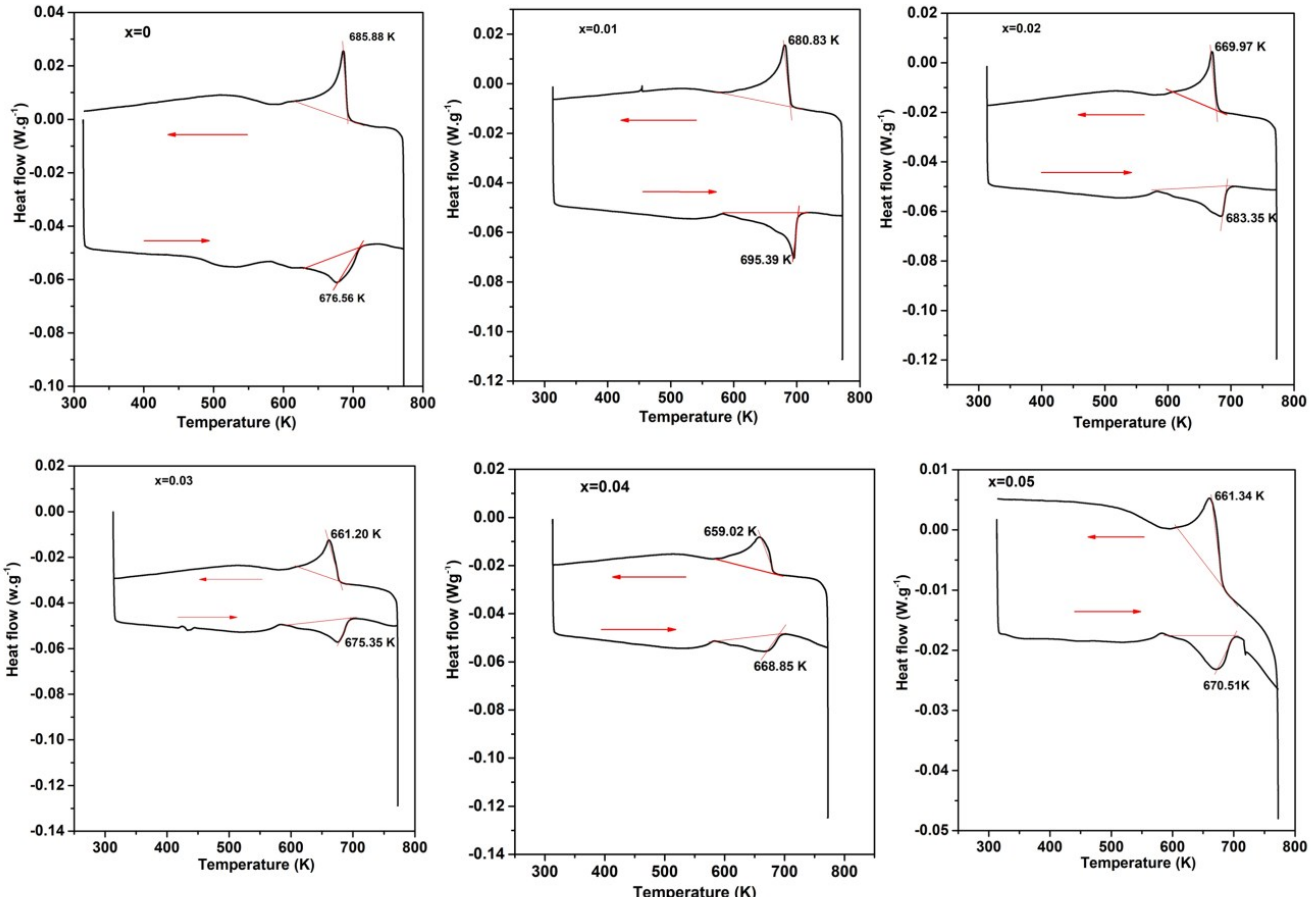

Figure S1. Heat flow from DSC measurements showing the phase transition temperature of $Ge_{1-x}Cd_xTe$ (x = 0-0.05).

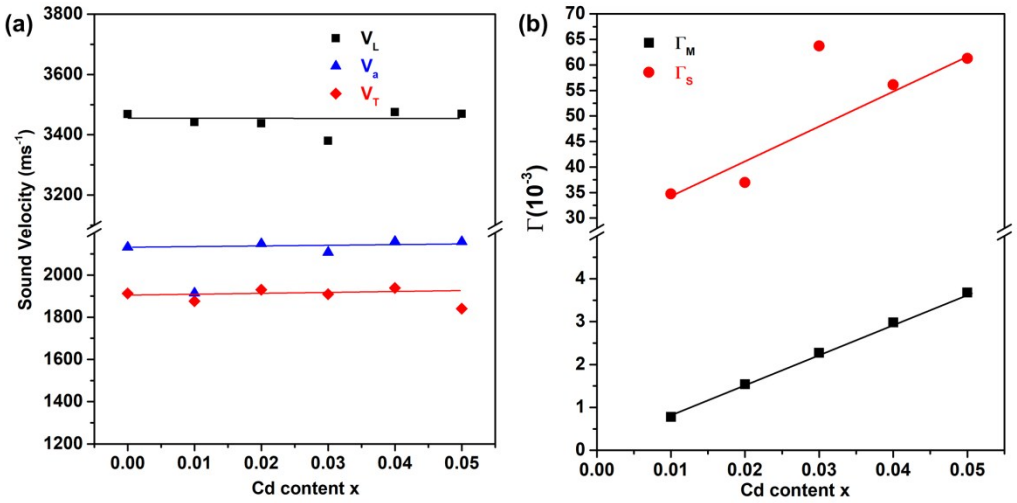

Figure S2. (a) Room temperature sound velocity and (b) mass fluctuation scattering parameter (Γ_M), and strain fluctuation scattering parameter (Γ_S) of $Ge_{1-x}Cd_xTe$ (x = 0-0.05).

Scattering parameter calculations: Callaway suggests that the impurity scattering parameter (Γ) can be calculated by fitting the lattice thermal conductivity of a disordered compound (κ_L) and the lattice thermal conductivity of an ordered pure GeTe (κ^P_L) compound through Eq. 1:

$$\frac{\kappa_L}{\kappa_L^P} = \frac{\tan^{-1} u}{u},\ u^2 = \frac{\pi^2 \Theta_D \Omega}{v^2} \kappa_L^P \Gamma \quad , \quad (1)$$

where u is the disorder scaling parameter, Θ_D is the Debye temperature (244 K for pure GeTe), Ω is the average atomic volume, h is the Planck's constant, and ν is the average sound velocity (2452 m/s for pure GeTe). In the model of Slack[1] and Abeles[2], considering the disorder scattering parameter as a combined value of disorder from both mass and strain field fluctuations allows to express Γ as: $\Gamma = \Gamma_M + \Gamma_S$, where Γ_M and Γ_S are the mass and strain fluctuation parameters, respectively, given by

$$\Gamma_M = \frac{\sum_{i=1}^{n} ci\left(\frac{\bar{M}_i}{\bar{\bar{M}}}\right)^2 f_i^1 f_i^2 \left(\frac{M_i^1 - M_i^2}{\bar{M}_i}\right)^2}{\sum_{i=1}^{n} ci}$$

(2)

$$\Gamma_S = \frac{\sum_{i=1}^{n} ci\left(\frac{\bar{M}_i}{\bar{\bar{M}}}\right)^2 f_i^1 f_i^2 \varepsilon_1 \left(\frac{r_i^1 - r_i^2}{\bar{r}_i}\right)^2}{\sum_{i=1}^{n} ci}$$

.

(3)

Here n is the number of different atoms in the lattice (n = 2 in GeTe) and ci is the degeneracy of the atomic occupancy ($c_1 = c_2 = 1$), $\bar{M}_i$ and $\bar{r}_i$ are the average atomic mass and radius on the i^{th} sublattice, respectively, $\bar{\bar{M}}$ is the average relative atomic mass of the compound, f_i^k is the fractional occupation of the k^{th} atoms on the i^{th} sublattice, ε_1 is the phenomenological adjustable parameter, M_i^k and r_i^k are the atomic mass and radius, respectively, expressed as:

$$M_i^k = \sum_k f_i^k M_i^k \tag{4}$$

$$r_i^k = \sum_k f_i^k r_i^k \tag{5}$$

$$\bar{\bar{M}} = \frac{\sum_{i=1}^{n} C_i \bar{M}_i}{\sum_{i=1}^{n} c_i}$$

.

(6)

Following the above expressions, a simplified expression for the impurity scattering parameter Γis derived and can be written as:

$$\Gamma = \frac{1}{4}\left(\frac{\bar{M}_1}{\bar{\bar{M}}}\right)^2 x(1-x)\left[\left(\frac{M_1^1 - M_1^2}{\bar{M}_1}\right)^2 + \varepsilon_1\left(\frac{r_1^1 - r_2^1}{\bar{r}_1}\right)^2\right] \quad (7)$$

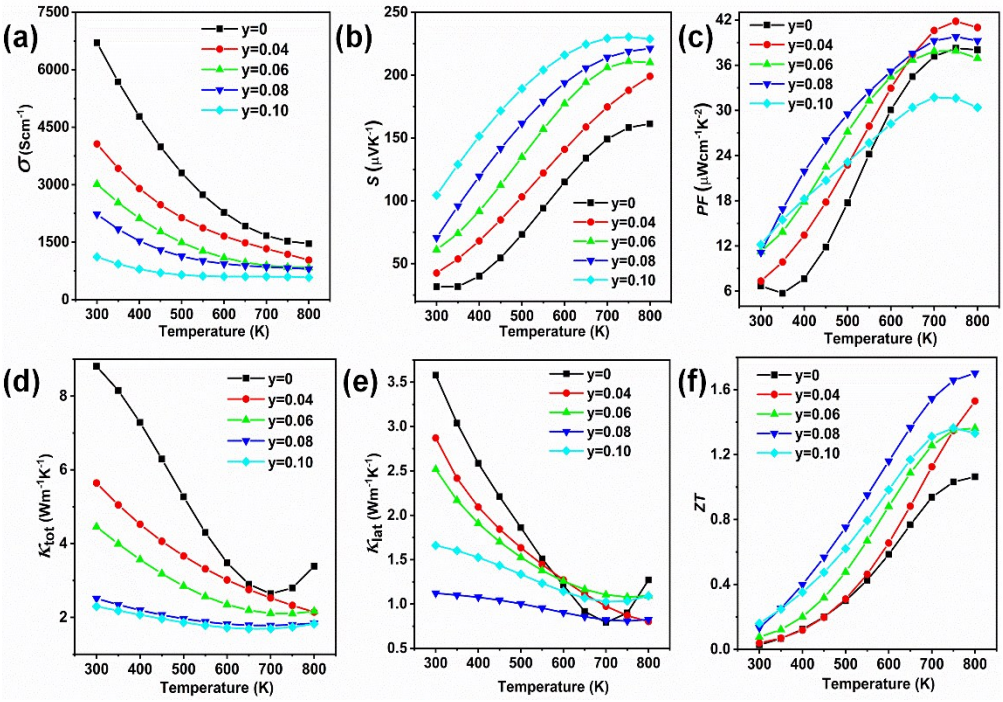

Figure S3. Temperature dependent thermoelectric transport properties for $Ge_{1-y}Sb_yTe$ (y = 0-0.10): (a) electrical conductivity, (b) Seebeck coefficient, (c) power factor, (d) total thermal conductivity, (e) lattice thermal conductivity and (f) figure of merit, *ZT*.

Electronic thermal conductivity and Lorenz number of $Ge_{0.97-y}Cd_{0.03}Sb_yTe$: The electronic thermal conductivity is calculated from the Wiedemann-Franz law, $\kappa_e = L\sigma T$, where L is the Lorenz number. L was calculated using the chemical potential, estimated by fitting the

experimental Seebeck coefficient. σ is the measured electrical conductivity and T is the absolute temperature.

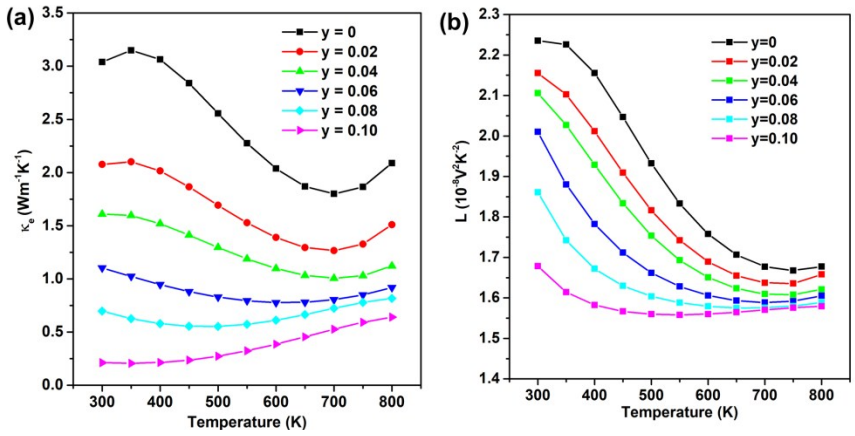

Figure S4. (a) Temperature-dependent electronic thermal conductivity of the $Ge_{0.97-y}Cd_{0.03}Sb_yTe$ (y = 0-0.10) sample and (b) Lorenz number as a function of temperature of $Ge_{0.97-y}Cd_{0.03}Sb_yTe$ (y = 0-0.10).

References

(1) Slack, G. A. Effect of isotopes on low-temperature thermal conductivity. *Physical Review* **1957,** *105* (3), 829.
(2) Abeles, B. Lattice thermal conductivity of disordered semiconductor alloys at high temperatures. *Physical Review* **1963,** *131* (5), 1906.